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OPTICAL PROPERTIES OF Nd DOPED RARE EARTH VANADATES (Preprint)

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14. ABSTRACT

Rare earth orthovanadates are being used as substitute for traditional solid state laser hosts such as yttrium aluminium garnet (YAG). While the most common of these is yttrium orthovanadate, other rare earth vanadates such as lutetium vanadate and gadolinium vanadate are being used for their special properties in certain applications. We report new measurements of the refractive indices and thermo-optics coefficients of these materials which will aid in the design of laser cavities and other nonlinear optical elements.

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Optical Properties of Nd Doped Rare Earth Vanadates

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Abstract

Rare earth orthovanadates are being used as substitute for traditional solid state laser hosts such as YAG. While the most common of these is yttrium orthovanadate, other rare earth vanadates such as lutetium vanadate and gadolinium vanadate are being used for their special properties in certain applications. We report new measurements of the refractive indices and thermo-optic coefficients of these materials which will aid in the design of laser cavities and other nonlinear optical elements.

Introduction

Investigation of rare earth vanadates as substitutes for traditional laser materials such as YAG is the result of their significant advantages as laser hosts[1-5].   Their higher absorption cross sections lead to lower lasing thresholds and their larger absorption bandwidth diminishes the frequency control requirements of the pump laser. Further, their large birefringence implies that the outputs of lasers made from these materials will be highly polarized. While most attention has been paid to YVO_4, and increasing number of studies are being devoted to $GdVO_4$ and $LuVO_4$ [6-13] due to important differences with YVO_4 which make them more attractive for

high power optical systems. For instance, $GdVO_4$ has a much larger thermal conductivity than YVO_4 [14,15] which reduces thermal management requirements at high powers. $LuVO_4$ has a much higher absorption cross section than YVO_4 at 880nm. The reduction of the wavelength shift between the absorption and emission wavelengths increases the slope efficiency of the laser and reduces the quantum defect resulting in less heating during high power operation. [16]

Design of intracavity components to control beam shape, and calculation of the modal behavior in guided wave lasers, optical resonator stability, and spectral characteristics of laser materials require precise values for the refractive indices and thermo-optic coefficients. The data available for both $LuVO_4$ and $GdVO_4$ for these materials is very limited both in spectral and temperature range [15, 17-19]. In this paper, we report a study of the refractive indices for $Nd:GdVO_4$ and $Nd:LuVO_4$ for wavelengths between .4 and 5.0 microns. Refractive indices were measured as a function of temperature between 22 and 225C at wavelengths from 0.4 to 1.3 microns. A standard Sellmeier equation was used to model the spectral behavior of the refractive indices and a second order polynomial for the thermo-optic coefficients was used to describe the temperature dependent behavior of the refractive index. These data were used to calculate the thermal lensing properties of the materials under specified pumping conditions and predict modifications in the calculation of oscillator strengths and transition rates derived from Judd-Ofelt theory.

Experiment

The prism samples used for this study were supplied by Coherent Crystal Associates and grown by the Czochralski method. The neodymium concentration was 0.5% for both materials. Both $GdVO_4$ and $LuVO_4$ are positive uniaxial crystals belonging to the tetragonal space group $I4_1/amd$. [20] The prisms were fabricated with the optic axis

perpendicular to the triangular faces of the prism. The apex angles of the prisms were measured

using an autocollimator and were $39.142^o \pm .001^o$ and $39.114^o \pm .001^o$ for GdVO$_4$ and LuVO$_4$

respectively.

The refractive index both as a function of wavelength and temperature was measured on a

Moller-Wedel divided circle spectrometer using the method of minimum deviation. The reader is

referred to ref [21, 22] for a complete description of the experimental protocol. This method is

still the most accurate way to measure the refractive index of materials. For thermo-optic

coefficient measurements, the advantage of the method is that it allows for direct measurement

of dn/dT without having to resort to the usual procedure of subtracting the contribution of the

thermal expansion coefficient from the overall change in optical path which is measured

interferometrically[23,24].   Room temperature refractive indices were measured at 43

wavelengths between .4046 microns and 5.0 microns. Refractive indices were measured at 9

temperatures between 22±1 C° and 225±1 C° at 9 different wavelengths. Standardization of the

experimental apparatus was done by comparing data obtained from calcium fluoride with those

of NIST[25,26]. Thermo-optic coefficients were within 5% of NIST data at all wavelengths and

refractive indices were within .0001. Analysis of the standard deviations of the angular

measurements coupled with accounting for the errors due to temperature fluctuations, gave us an

estimated error in the refractive index of approximately $1.5X10^{-4}$.

Results

The room temperature refractive indices of GdVO$_4$ and LuVO$_4$ are shown in figure 1. A

Levenburg-Marquardt algorithm was used to fit a three term, 5- parameter Sellmeier equation of

the form

$$n^2 = A + \frac{B\lambda^2}{\lambda^2 - C} + \frac{D\lambda^2}{\lambda^2 - E}$$

Table 1 shows the calculated Sellmeier coefficients for the two materials. A comparison

between the data and the Sellmeier equations shows that the deviation between the model and the

data is less than $2X10^{-4}$. The values for the refractive indices vs. temperature are shown in fig. 2.

While the index appears to increase linearly with temperature yielding a temperature independent

value for dn/dT, closer examination reveals a small curvature in the n vs. T data similar to that

found in YVO_4[22]. To approximate the temperature dependence, the data were fit to a second

order polynomial of the form

$$n = n(23) + A(T - 23) + B(T - 23)^2$$

Values for dn/dT as a function of temperature were obtained by differentiating the resulting

expression, resulting in dn/dT=A+2B (T-23). The values for A and B are shown in Table 2.

Statistical analysis shows that this model is somewhat more likely than the linear model in the

visible spectrum but it is unclear whether the parabolic model is justified at wavelengths longer

than 650 nm.

Discussion

The utility of a given material as a laser source is determined by a number of physical

characteristics including thermal conductivity, thermo-optic behavior and spectroscopic

characteristics and accurate determination of the refractive indices and thermo-optic coefficients

of these materials is critical to the prediction of their behavior in optical systems.   Regarding

spectroscopic characteristics, the work of Judd [27] and Ofelt [28], modified by several other

researchers [29-33] has provided the best means of determining oscillator strengths and

radiation transition rates for rare earth ions embedded in host materials. According to Judd-Ofelt

theory [33], the oscillator strength may be written as

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$$f = \frac{8\pi^2 mc}{h\lambda e^2} n \left(\frac{n^2 + 2}{3n}\right)^2 \sum_{MM'} \left| \left\langle \alpha JM \left| P \right| \alpha'J'M' \right\rangle \right|^2$$

where n is the refractive index, P is the electric dipole transition operator and $\left\langle \alpha JM \right|$ and

$\left| \alpha'J'M' \right\rangle$ are the initial and final states for the dipole transition.

The dependence of the oscillator strength on the refractive index means that the errors in its

calculation may be written as

$$\Delta f = \frac{\partial f}{\partial n} \Delta f = \frac{3n^4 + 4n^2 - 4}{n^5 + 4n^3 + 4n} \Delta n$$

.In the case of LuVO$_4$, the refractive indices are 1.5% smaller than those for YVO$_4$, which were

used in [16]. This results in an error of 6%-8% in the calculated strength of the oscillators. For

the case of GdVO$_4$, the differences will be between 1% and 5%.

Thermal lensing is another critical problem which needs to be considered for high power

lasers. Nonuniform changes in the optical path length caused by thermal expansion due to

inhomogeneous laser beams and temperature dependence of the refractive index result in

unacceptable beam distortions in the far field. The focal lengths in samples of GdVO$_4$ have been

measured as a function of input power [34]. No similar measurements have been made on

LuVO$_4$ as the necessary data is nearly nonexistent. The focal length of a cylindrical laser rod

with a given end pump power is [35]

$$f = \frac{\pi K_c \, \omega_p^2}{P_{ph}\left(\frac{dn}{dT}\right)} \left(\frac{1}{1 - \exp(-\alpha l)}\right)$$

where $\frac{dn}{dT}$ is the change of refractive index with respect to temperature, P_{ph} is the fraction of the

pump power contributing to the heating of the crystal in watts, α is the absorption coefficient, K$_C$

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is the thermal conductivity, l is the length of the laser rod and ω_p is the spot size of the input laser. We assume that αl is large enough so that we may ignore the exponential term and we further assume that P_{ph} and ω_p are the same for all the materials. The differences in focal length of the materials are then due solely to the differences in dn/dT and the thermal conductivity. For the purpose of the calculations, we considered a 400 micron spot size, and used values from the literature for the thermal conductivity[15]. The result of this calculation is shown in figure 5, where the relative focal length is plotted vs. input power for both $LuVO_4$ and $GdVO_4$. The focal length for YVO_4 is also given for comparison. It is clear that due to their larger thermal conductivity and thermo-optic coefficient that $GdVO_4$ and $LuVO_4$ have lower thermal focusing power than YVO_4. That said, it is also clear that these values exceed considerably those calculated by Liu [34]. We make note of the fact that the values of the thermo-optic coefficients used in ref [34] are much smaller than those we measured in our laboratory. Given the careful standardization done for our experiments, we suggest that some of the assumptions used in developing the measurements of the focal power may not be valid.

Conclusion

We have measured the refractive indices and thermo-optic coefficients of $GdVO_4$ and $LuVO_4$ over a wide range of wavelengths and temperatures. Results show that these materials provide somewhat better thermal lensing properties than YVO_4 and the results may be used to modify predictions made by Judd-Ofelt theory.

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Table 1. Sellmeier Coefficients for .5% $Nd:GdVO_4$ and .5% $LuVO_4$

Sellmeier Coefficient	$GdVO_4$ n_o	$GdVO_4$ n_e	$LuVO_4$ n_o	$LuVO_4$ n_e
A	2.394	2.7864	2.4617	2.8835
B	1.4394	1.9314	1.4546	1.8623
C	0.0493	0.0573	0.0505	0.0586
D	1.7046	2.7778	1.7781	2.9081
E	165.1	182.81	162.27	187.18

Table 2. Parameters for determining thermo-optic coefficients .5% Nd doped GdVO$_4$

Wavelength (Microns)	GdVO$_4$ no A x10^{-5}	GdVO$_4$ no B x10^{-8}	GdVO$_4$ ne A x10^{-5}	GdVO$_4$ ne B x10^{-8}	LuVO$_4$ no A x10^{-5}	LuVO$_4$ no B x10^{-8}	LuVO$_4$ ne A x10^{-5}	LuVO$_4$ ne B x10^{-8}
.4046	3.822	3.240	3.8569	5.4843	3.9377	3.9395	3.8599	5.1195
.45	2.7406	3.1302	2.7109	2.9595	3.00	2.3614	2.5709	2.8078
.5	2.7152	0.6760	2.1635	2.2410	2.5109	1.9272	2.0310	1.7979
.55	2.2158	1.5376	1.6234	2.8960	2.0655	2.7056	1.5822	1.8734
.6	1.9010	1.4542	1.4240	1.9771	2.0033	1.6192	1.4049	1.1622
.65	1.6837	1.6614	1.2545	2.1808	1.9148	1.2201	1.025	3.4806
.7	1.9583	-0.554	1.06	1.01	2.0429	0.37967	1.2885	0.04943
.9	1.6784	-0.2423	0.761	0.950	1.6701	0.79707	0.90694	-0.2139
1.1	1.1798	2.9658	0.900	-0.02	1.5634	0.87996	0.67586	2.3026
1.3	1.1012	2.6256	0.720	0.309	1.4884	0.95457	0.68482	1.9202

Table 3. Values used for the focal length calculations

Parameter	YVO$_4$	GdVO$_4$	LuVO$_4$
Absorption coefficient cm$^{(-1)}$	31.4 @808nm [brochure]	~45?@808nm (π pol) ~37@808nm (σ pol) [Higuchi,JCG264, 284	60@808nm (π) 18@808nm (σ) (Higuchi, JCG283, 100 (2005)
Thermal Conductivity W-m/^{o}C	5.23 ($\parallel$c) 5.10 ($\perp$ c) [brochure]	11.4 (c) 10.1 (a)	9.94 ($\parallel$c) 8.14 ($\parallel$a) [Cheng, Appl Phys B 86, 681 (2007)

Figure Captions:

Figure 1. Refractive Indices of Nd:GdVO$_4$

Figure 2. Refractive Indices of Nd:LuVO$_4$

Figure 3a. Ordinary Refractive Index of GdVO4 vs. Temperature, Visible spectrum

Figure 3b. Ordinary Refractive Index of GdVO4 vs. Temperature, Near infrared spectrum

Figure 3c. Extraordinary Refractive Index of GdVO4 vs. Temperature, Visible spectrum

Figure 3d.   Extraordinary Refractive Index of GdVO4 vs. Temperature, NIR spectrum

Figure 4a. Ordinary Refractive Index of LuVO4 vs. Temperature, Visible spectrum

Figure 4b. Ordinary Refractive Index of LudVO4 vs. Temperature, Near infrared spectrum

Figure 4c. Extraordinary Refractive Index of LuVO4 vs. Temperature, Visible spectrum

Figure 4d.   Extraordinary Refractive Index of LuVO4 vs. Temperature, NIR spectrum

Figure 5. Thermal focal power vs. Input power
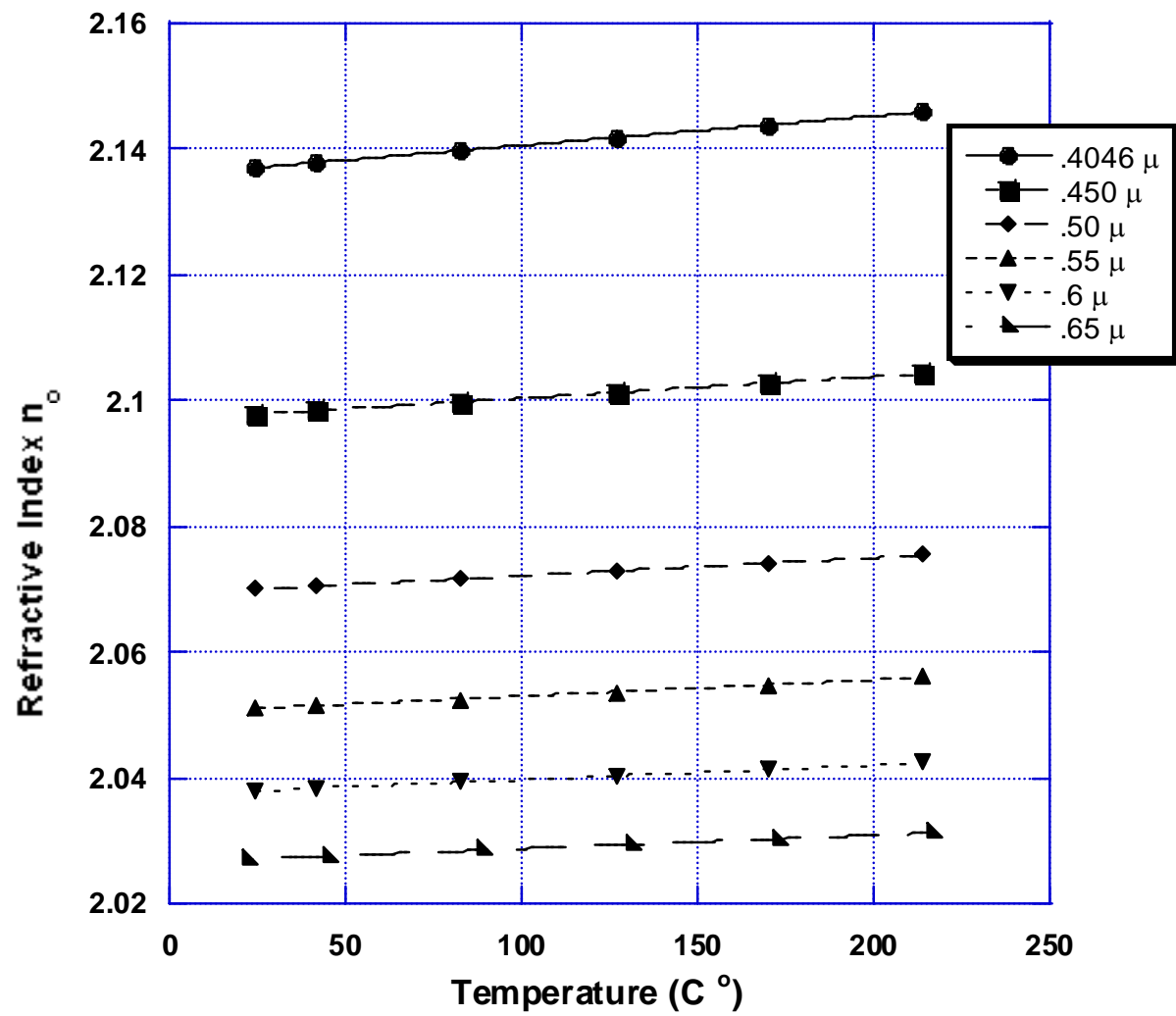
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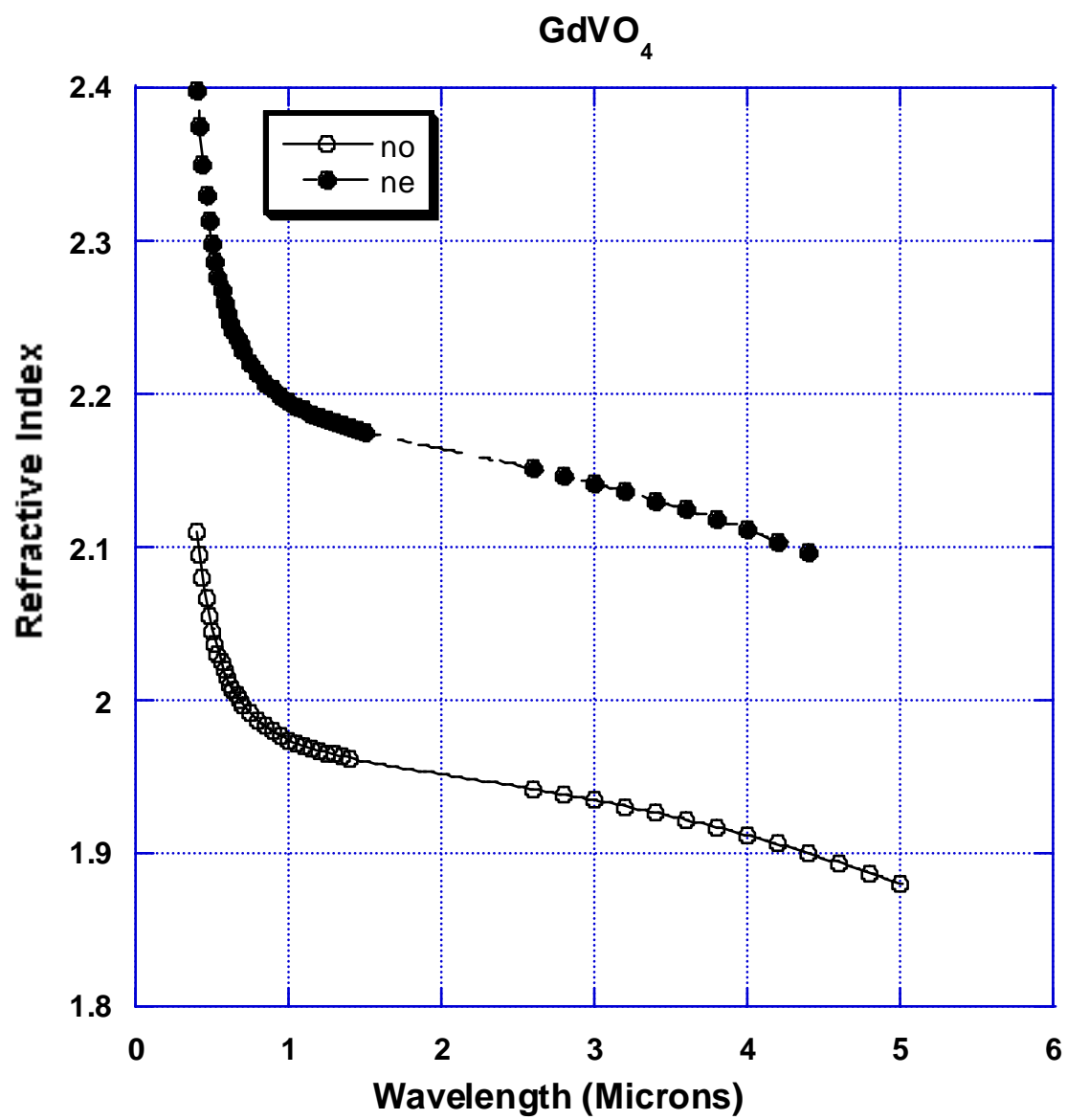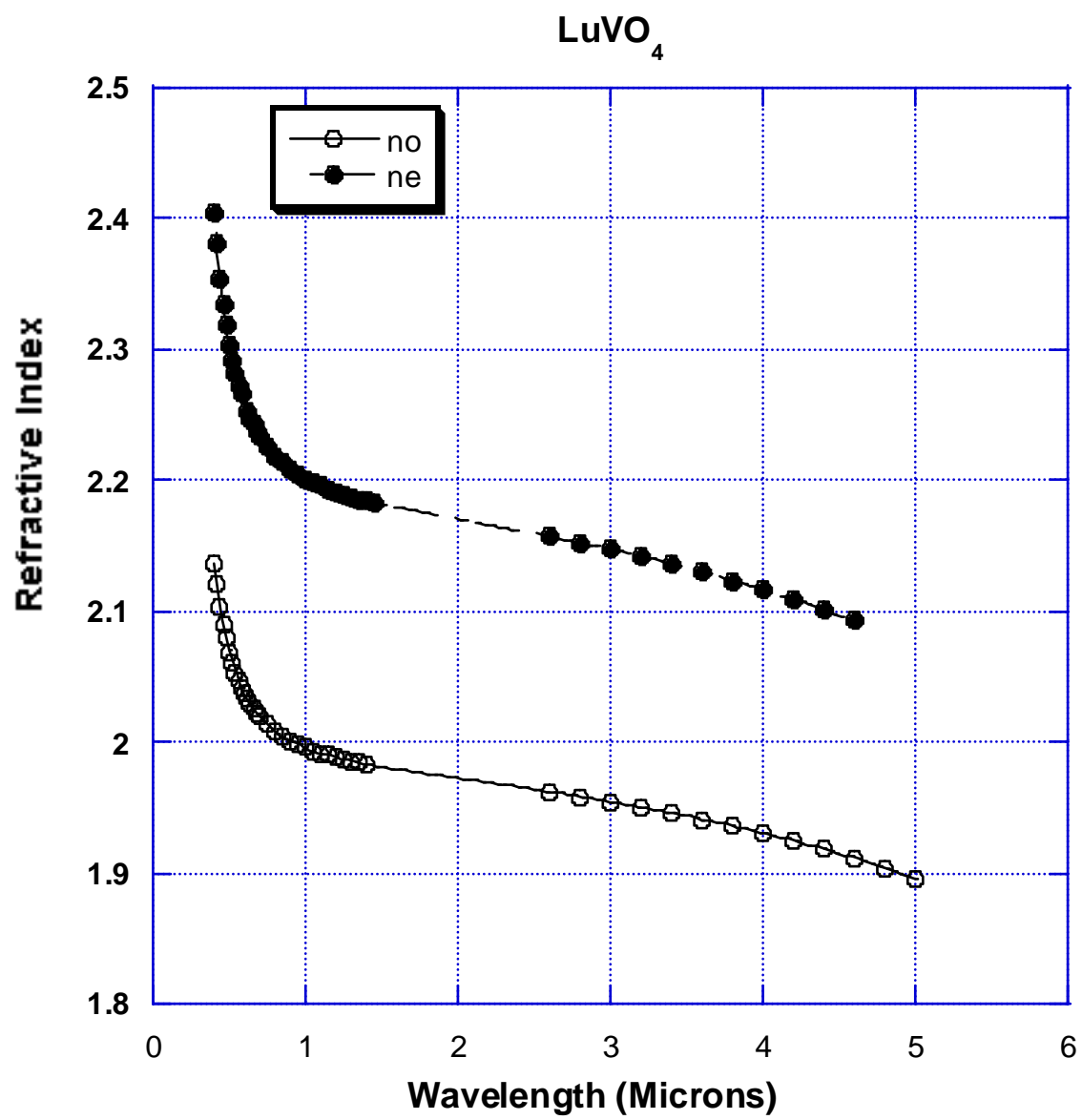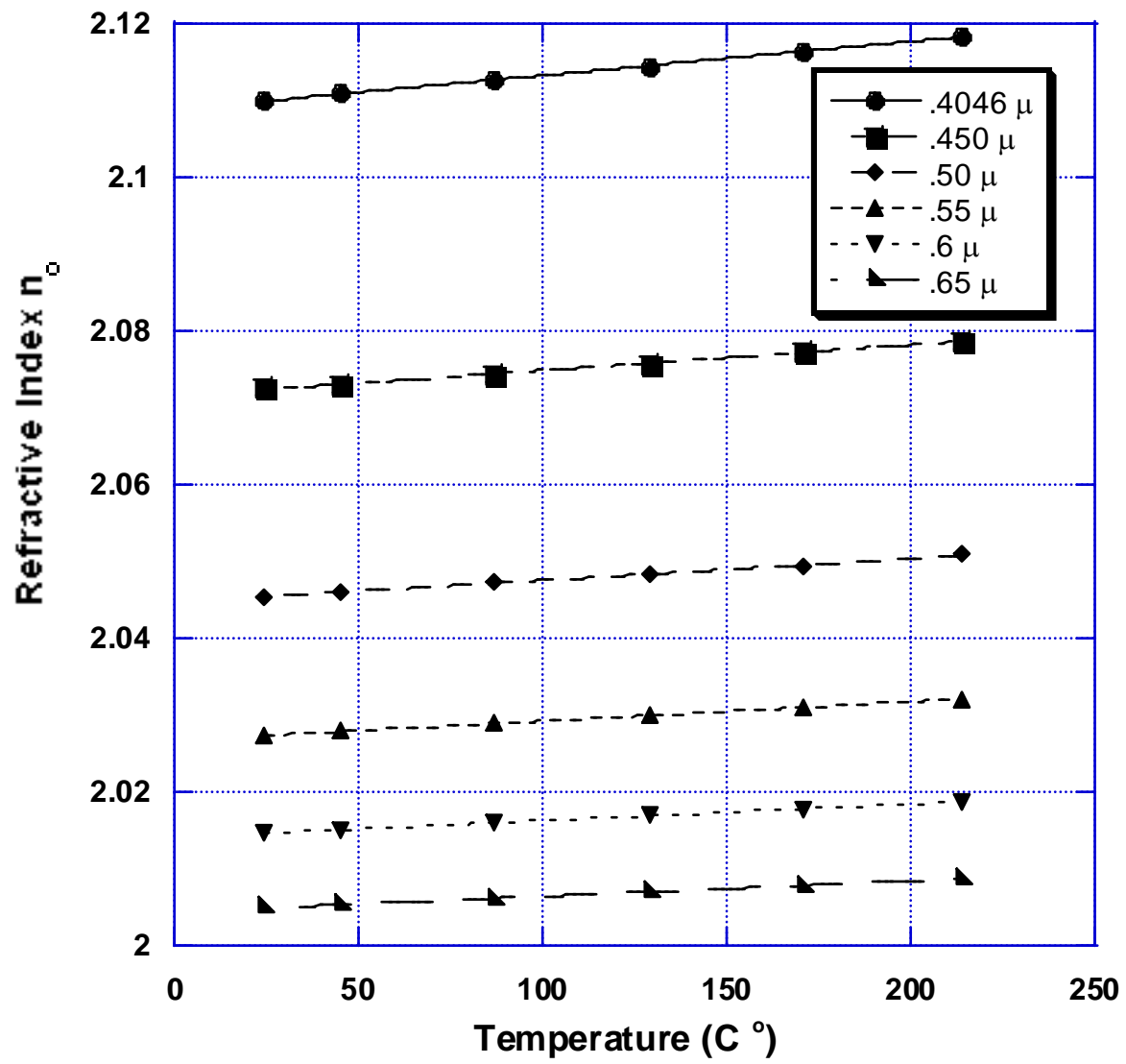
Figure 1

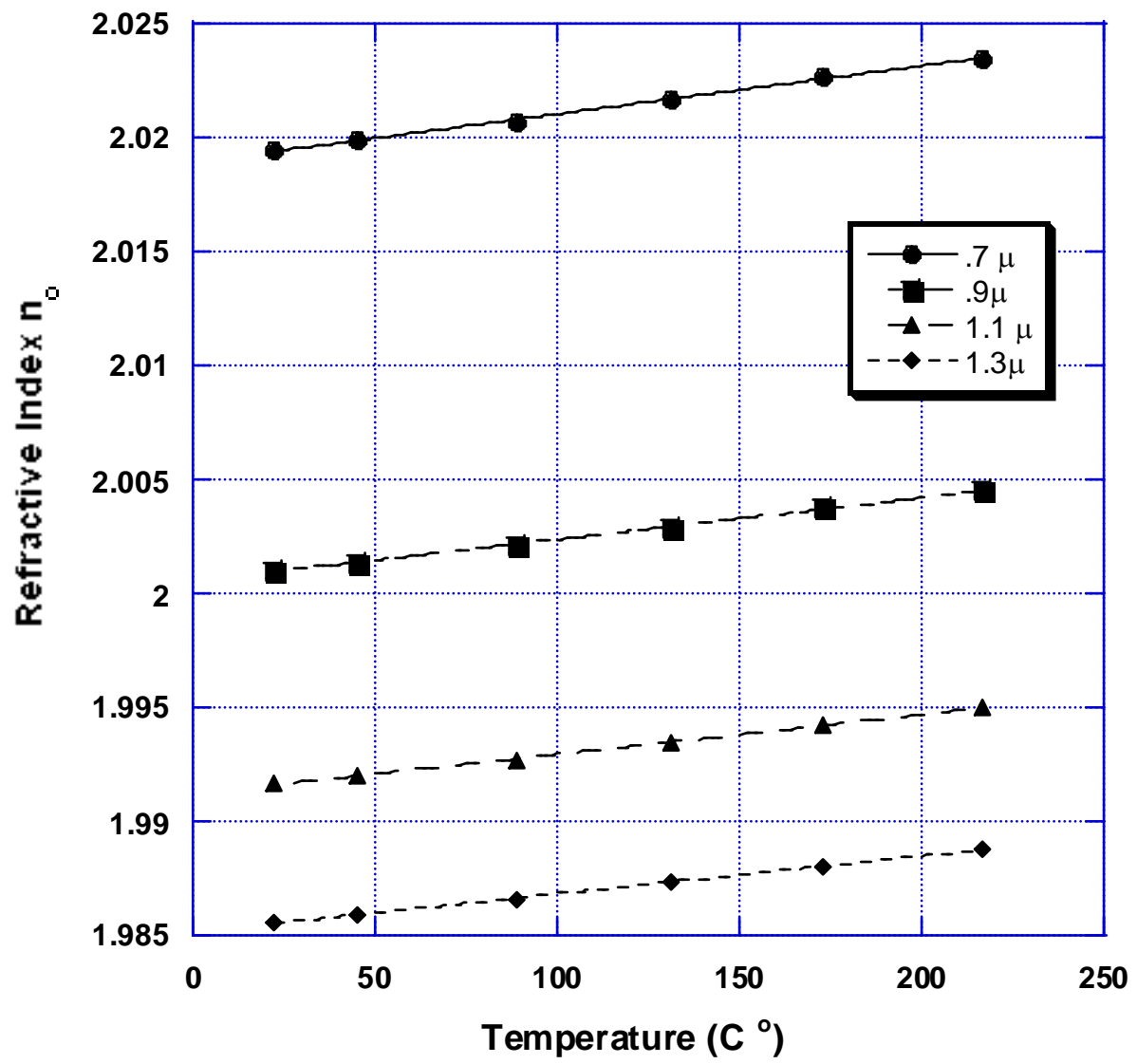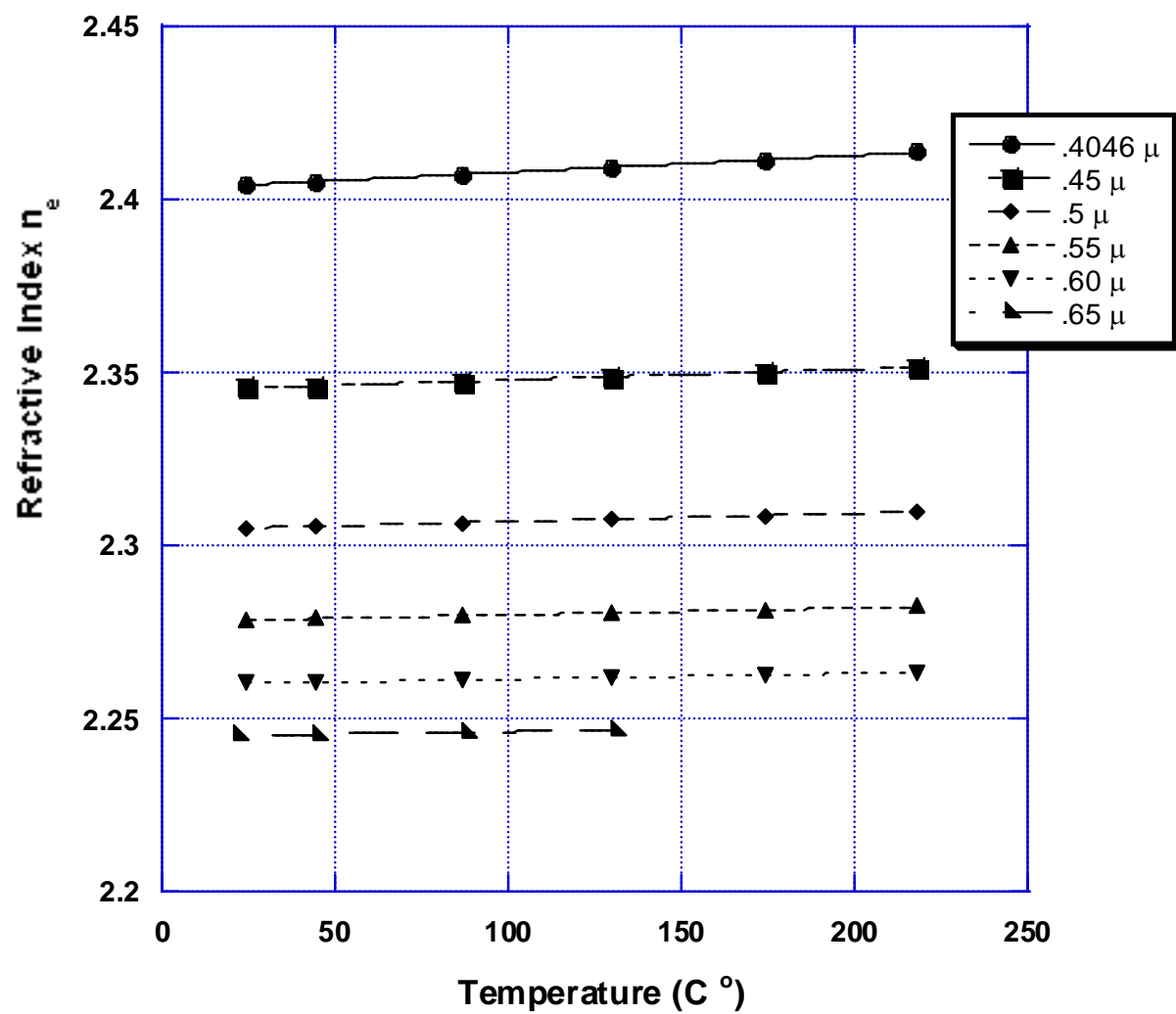Figure 2

Figure 3a.
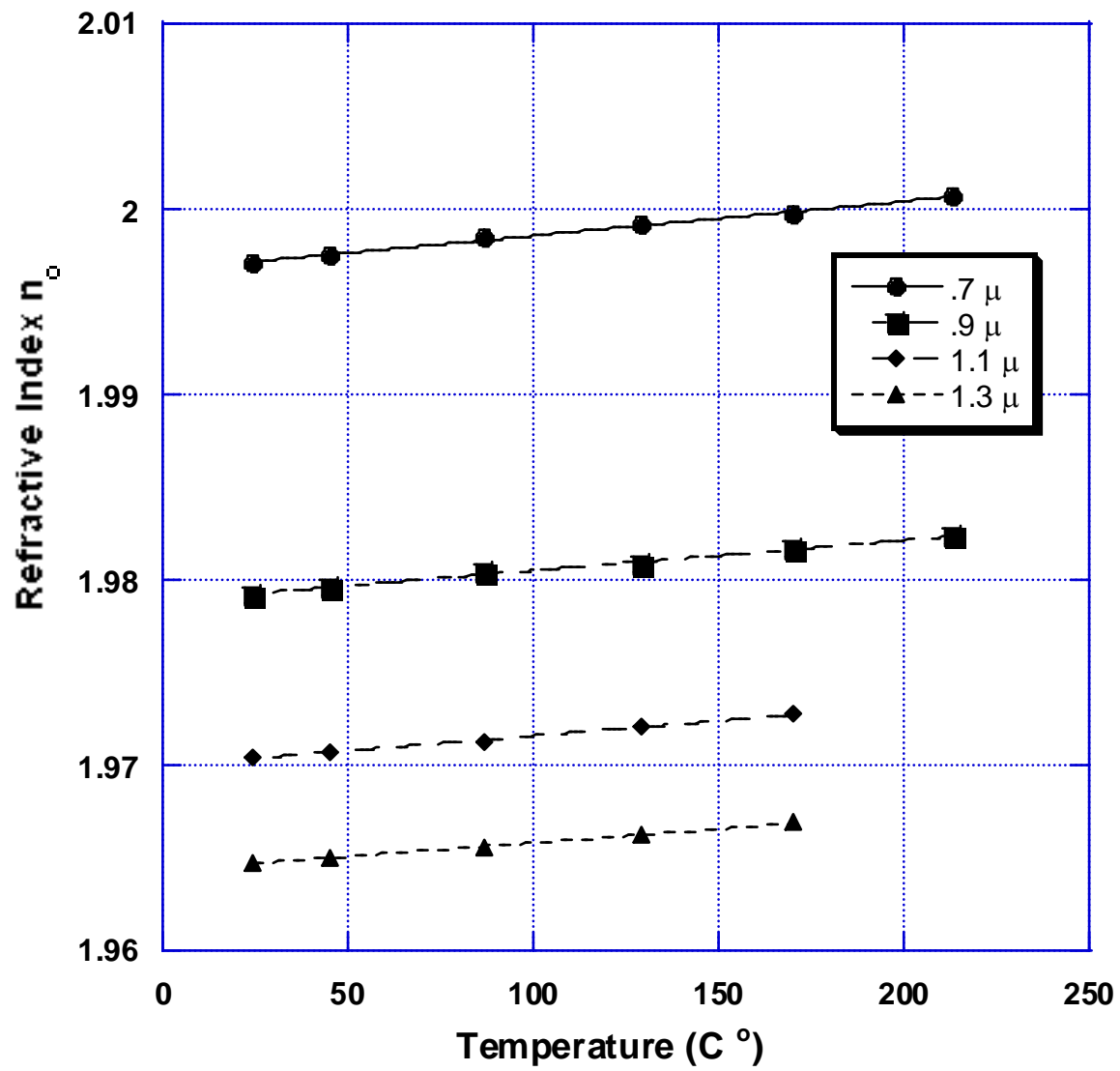
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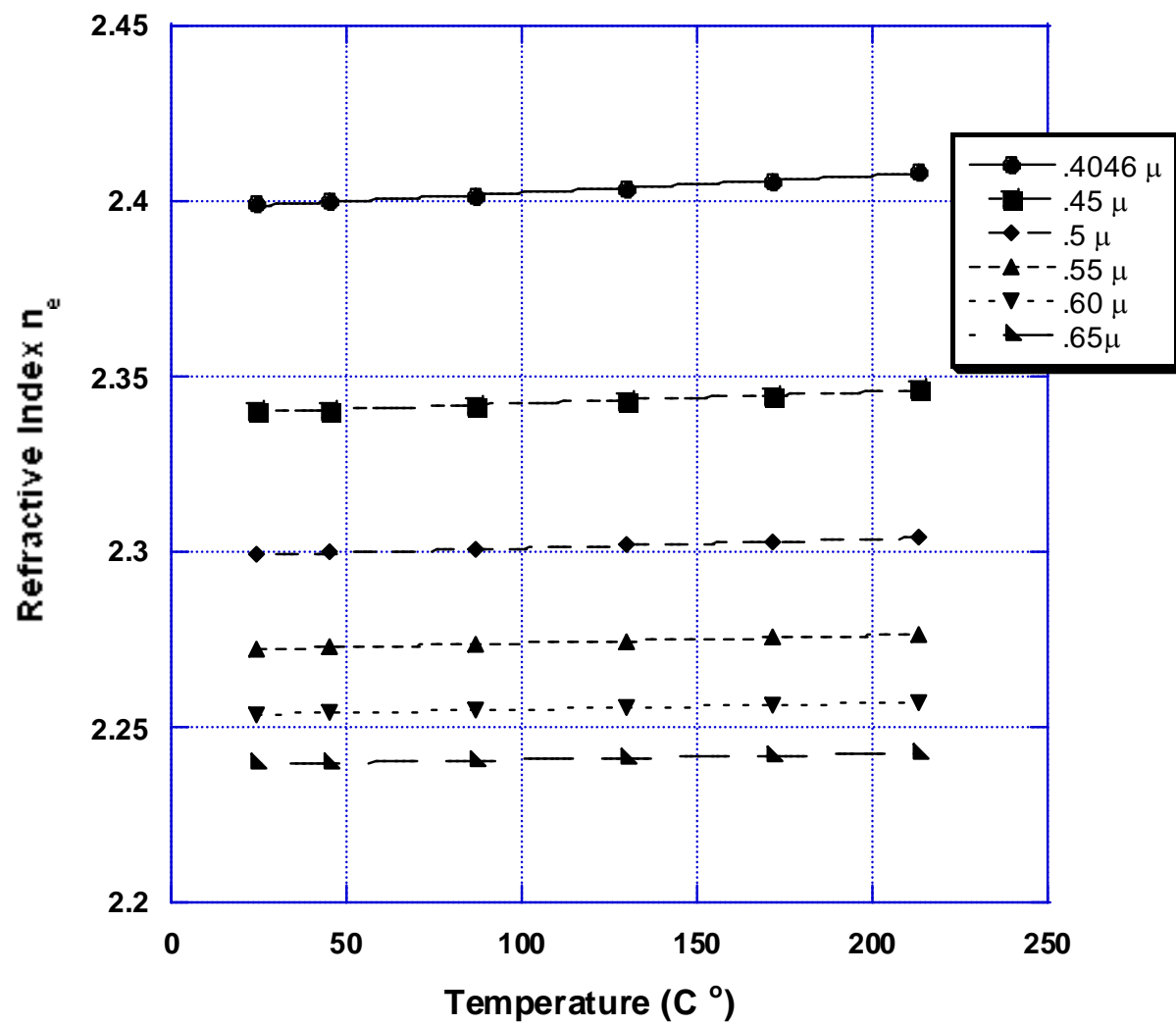
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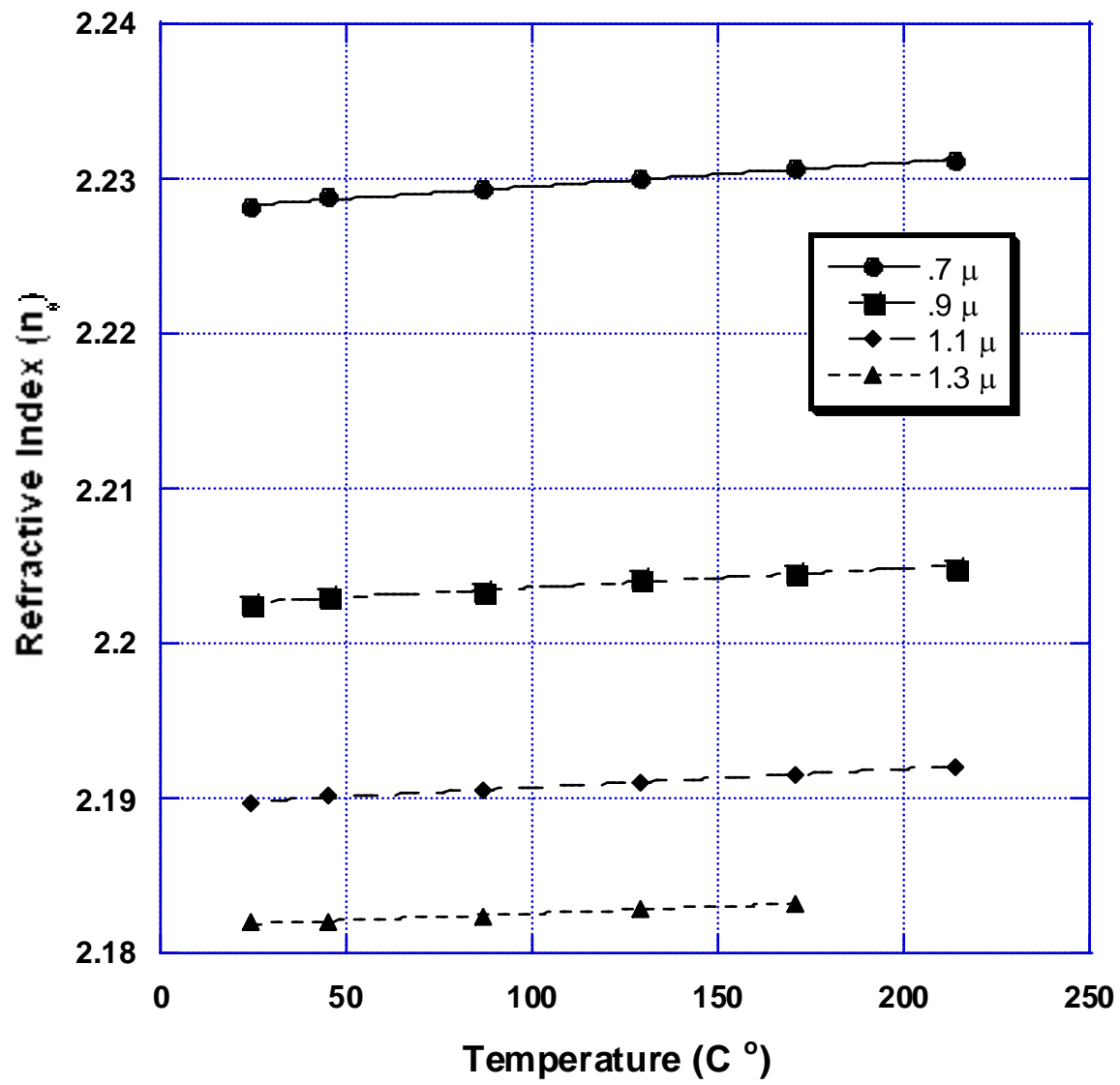
Figure 3d.

Figure 4a.

Figure 4b.

Figure 4c.

Figure 4d.

Figure 5.